Chin. Geogra. Sci. 2020 Vol. 30 No. 5 pp. 921–933
https://doi.org/10.1007/s11769-020-1154-8

Springer Science Press
www.springerlink.com/content/1002-0063

A High Resolution Emission Inventory of Domestic Burning in Rural Region of Northeast China Based on Household Consumption

ZHAO Hongmei[1], YANG Guangyi[1, 2], XIU Aijun[1], ZHANG Xuelei[1]

(1. *Key Laboratory of Wetland Ecology and Environment*, *Northeast Institute of Geography and Agroecology*, *Chinese Academy of Sciences*, *Changchun* 130102, *China*; 2. *University of Chinese Academy of Science*, *Beijing* 100049, *China*)

Abstract: Domestic burning emits large amounts of pollutants into the ambient air due to incomplete and inefficient combustion, with significant impacts on indoor air quality and human health. Northeast China is one of the major contributors to domestic burning emissions in China; however, the high-resolution emissions inventories of domestic biomass and coal burning in Northeast China are lacked, which are needed to estimate the extent of its impact. In this study, we established a town-level emissions inventory of gaseous pollutants and particulate matter (PM) from domestic biomass and coal burning, based on per household consumption in each town in rural region of Northeast China. The results revealed that biomass burning was the major domestic burning source over the region in 2016. Domestic biomass burning is the major contributor to PM and volatile organic compounds (VOCs) emissions, while coal burning is the major contributor to SO_2 emissions. High emissions intensities were concentrated around the cities of Harbin, Suihua, Changchun, Qiqihar, and Chifeng, each with emissions intensity for $PM_{2.5}$ and VOCs of more than 2000 Mg per 27 km × 27 km grid cell. Additionally, there are three burning peaks (6−7 am, 12 pm and 4−7 pm) during both the heating (from October to April) and non-heating seasons (from May to September), due to cooking and heating. The burning peaks in the non-heating season were more notable than those in the heating season. These results suggest that the government should pay more attention to domestic biomass and coal burning in rural areas, in order to reduce pollutant emissions and control regional haze during the heating season.

Keywords: residential emission; particulate matter (PM); source contribution; indoor air pollution; spatiotemporal distribution

Citation: ZHAO Hongmei, YANG Guangyi, XIU Aijun, ZHANG Xuelei, 2020. A High Resolution Emission Inventory of Domestic Burning in Rural Region of Northeast China Based on Household Consumption. *Chinese Geographical Science*, 30(5): 921–933. https://doi.org/10.1007/s11769-020-1154-8

1 Introduction

Solid fuels (e.g. biomass, coal) burning contributes large quantities of gaseous pollutants and particulate matters to the atmosphere, having a significant impact on air quality, human health, and climate change (Chen et al., 2017; Yadav et al., 2017). Globally, approximate 2.8 billion people rely on solid fuels (Bonjour et al., 2013). It estimated that about 50% of all households and 90% of rural household use solid fuels for cooking and heating at the global scale (Desai et al., 2004). Domestic solid fuels burning in the households of the developing countries is estimated to be one of the main health risk worldwide (Mestl et al., 2007). Pollutants with fine particles penetrate deeply into human body, including lungs and blood, and has been identified as the most serious air pollutant for human health by the World Health Organization (WHO). About 3.8 million people a year died prematurely from illness attributable to the household air pollution caused by the inefficient use of solid fuels

Received date: 2019-10-30; accepted date: 2020-01-09

Foundation item: Under the auspices of National Key Research and Development Program of China (No. 2017YFC0212303, 2017YFC0212304), National Natural Science Foundation of China (No. 41771504), National Natural Science Foundation of Jilin Province (No. 20200201214JC)

Corresponding author: ZHAO Hongmei. E-mail: zhaohongmei@iga.ac.cn

and kerosene for cooking (WHO, 2018). As important kinds of solid fuels, biomass and coal burning have significant impacts on regional air quality and human health.

Biomass burning is one of the primary source of $PM_{2.5}$ (particles with an aerodynamic diameter of 2.5 μm or less). Of this, domestic biomass burning is one of the major sources, contributing 57% of the total biomass burning emissions in China for all pollutants (Zhou et al., 2017). Compared with open field burning, domestic burning is characterized by incomplete and inefficient combustion without air pollution control devices, and, therefore, more negatively impacts indoor air quality and human health (Cheng et al., 2017; Li et al., 2017). It has been estimated that about 36% of primary $PM_{2.5}$ and 53% of elemental carbon (EC) of China's annual anthropogenic emissions have been due to domestic solid fuels combustion (Wang et al., 2012; Shen et al., 2012; Huang et al., 2014). Especially in rural regions of China, biomass and coal are major sources of energy for heating and cooking (Li et al., 2017), which is important when considering that rural cooks spend more hours cooking and create emissions with $PM_{2.5}$ concentrations 5.4 times greater than urban cooks (Jiang and Bell, 2008). Therefore, domestic burning in the rural region have significant impacts on indoor air quality and human health.

Domestic burning emission have been conducted all over the world, particularly in Europe and USA (Badamassi et al., 2017; Wilnhammer et al., 2017). However, the research in developing country is limited (Fleming et al., 2018). In China, pollutant emissions from solid fuels represent a major and under acknowledged anthropogenic source of air pollution (Liu et al., 2016). Previous studies have mainly focused on emission factors (EF) and characteristics of the many kinds of gaseous pollutants and particulate matters (Shen et al., 2012; Shen et al., 2015; Li et al., 2016), as well as the impacts of domestic burning emissions on household air quality and human health (Zhang and Smith, 2007; Badamassi et al., 2017; Li et al., 2017; Du et al., 2018). To our knowledge, high-resolution inventories of domestic burning emissions, with hourly temporal and sufficient spatial sampling, are limited. This is often compounded by a lack of key information, such as the amount of consumption, burning time and burning duration, which are key parameters for the estimation of fuel consumption. The emissions inventory contains the foundational data for estimating the impact of indoor air quality on human health, and controlling domestic air pollution. Conversely, over recent years in particular, the Chinese government has focused its attempts to reduce severe air pollution on major pollution sources such as vehicles and power plants. With domestic burning emissions poorly understood and managed, it is possible that they contribute far more to overall air pollution in the region than has been previously acknowledged (Liu et al., 2016).

It is well known that air pollution dominated by $PM_{2.5}$ in China is severe. To control the air pollution, the emission inventory of $PM_{2.5}$ and other pollutants need to be calculated first. Domestic burning as one of the primacy source of $PM_{2.5}$, its emission inventory is necessary to be known. It is estimated that approximate 43% of investigated households use biomass and coal for daily cooking and 29% of them rely on biomass and coal for heating in China in 2012 (Duan et al., 2014). In rural households, the ratio of biomass and coal used for daily cooking was about 47.6% and 13.5%, particular in Northeast China, biomass account for more than half of the total household fuel-use (Duan et al., 2014). However, the high resolution of domestic burning emission in rural region of Northeast China is unknown. In fact, Northeast China is one of the major contributing regions to domestic biomass burning emissions (Tian et al., 2017; Xing et al., 2018). At the same time, Northeast China has experienced severe regional haze in recent years, due to quality of pollutants emissions form crop residue burning and coal consumption for heating (Chen et al., 2018a; Wen et al., 2018; Li et al., 2019b; Yang et al., 2020). Compared with other regions, such as the Beijing-Tianjin-Hebei region, the Yangtze River Delta region, and the Pearl River Delta region, efforts to control air pollution in Northeast China is not well effective. It is urgent to reduce $PM_{2.5}$ concentrations and eliminate regional heavy haze events in Northeast China. High resolution emission inventory need to be calculated first to eliminate air pollution, particular for the key emission source (e.g., biomass burning, coal burning for heating). Therefore, in this study, we calculate the total emissions from domestic burning for each town of Northeast China based on amount of fuel burnt in every household, and analyze the spatial and temporal distribution of these emissions. The study is meaningful for understanding the spatiotemporal characteristic of anthropogenic sources and underlying mechanisms of regional haze events. The results would help to identify key emission sources, target the pollution affecting their communities, and provide scientific references for policy making to control air pollution in this region.

2 Study Area and Methodology

2.1 Study area

The Northeast China consists of Liaoning, Jilin and Heilongjiang provinces, and four prefectural-level cities in the eastern region of Inner Mongolia Autonomous Region (i.e., Tongliao City, Chifeng City, Hulun Buir City, and Hinggan League) (Fig. 1). The Northeast China is one of the major grain producing regions in China, the planting area is mainly concentrated in the Songnen Plain and Sanjiang Plain. The planting pattern is single cropping, conventional planting in May and harvesting in earlier October. The main crop types are corn, rice, soybean, and wheat. The non-growth season is from late October to the following Aril, due to the cold climate. The annual mean temperature is 4.7℃, and in January the monthly mean temperature is −12.8℃. Therefore, domestic heating is concentrated in the months of October to April (lasting for six months), which is about one month longer than that the heating

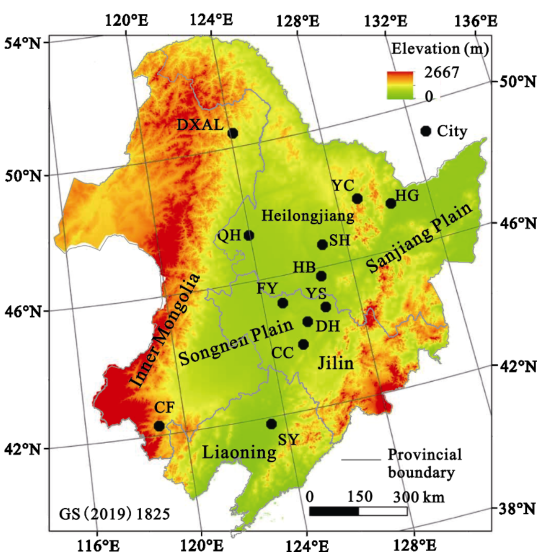

Fig. 1 The location of Northeast China and some high emission regions mentioned in this study. SY, CC, HB, SH, FY, DH, YS, QH, CF, HG, YC, and DXAL represent Shenyang, Changchun, Harbin, Suihua, Fuyu, Dehui, Yushu, Qiqihar, Chifeng, Hegang, Yichun, and Da Hinggan Ling, respectively

season in Beijing, North China (Li et al., 2019b). Over the heating season, the uncontrolled and inefficient combustion of solid fuels means that every household can emit large amounts of pollutants into the ambient air (Liu et al., 2016; Li et al., 2017; Chen et al., 2018b). Especially in the rural region of Northeast China, heating is different with the centralized heating in city, people usually heating in every household. The major solid fuels used are biomass and coal; the main types of biomass used in rural areas are wood and crop residue, and the main coal types are bituminite, briquette and anthracite.

2.2 Data source and methodology

2.2.1 Description of domestic burning emissions inventory

In this study, we developed a town-level domestic biomass burning inventory by compiling statistical and survey data. The annual domestic burning emissions were calculated using the following Equation (Zhou et al., 2017):

$$E = EF \times M \tag{1}$$

where E is the amount of a particular pollutant emitted (Mg), EF is the emission factor (g/kg) (a detailed description of EF is presented in section 2.2.2), and M is the amount of biomass or coal burnt (Mg).

2.2.2 Determination of emission factors

Emission factors (EF) are important parameters in calculating emissions inventories. To improve the accuracy of the emissions inventory, the following process was followed to select the emission factors. For domestic biomass burning, we used EFs published from laboratory experiments, where they were available. If there were no laboratory data, localized open field burning data were selected. For domestic coal burning, emission factors were collected from technical guides used to calculate air pollutant emission inventories due to domestic coal burning. All EFs used in this paper were shown in Table 1.

2.2.3 Determination of activity data

In this study, we developed a new method to collect the activity data, using the amount of fuel burnt every household and the number of households in every town. In December 2017, we designed paper questionnaire to collect activity data and the survey was conducted in local university. The students whose family in the rural

Table 1 Emission factors (g/kg) of particulate matter and gaseous pollutants from published papers used in this paper for domestic biomass and coal burning

Fuel type	SO_2	NO_x	PM_{10}	$PM_{2.5}$	VOC_S	NH_3	CO	EC	OC	CO_2	CH_4
Rice	0.48[h]	1.92[a,b,d,f]	6.88[h]	6.40[h]	8.40[h]	0.52[h]	79.70[a,b,h]	0.49[a]	2.01[a]	1147.40[a.b,d]	4.80[b]
Corn	1.33[h]	1.86[a,b,h]	7.39[h]	6.87[h]	7.34[h]	0.68[h]	82.37[a,b,c,d,h]	0.95[a]	2.25[a]	1491.00[d]	3.91[c]
Soybean	0.53[e,h]	1.12[e,f,g]	7.69[h]	7.15[h]	8.82[b,i]	1.30[b,e]	80.70[f]	0.51[e,g]	2.21[e,c]	963.40[b]	6.08[b]
Bituminite	7.40[j]	1.60[j]	13.50[j]	10.80[j]	4.00[j]	–	140.10[j]	5.00[k]	3.40[k]	–	–
Briquette	6.80[j]	0.80[j]	1.10[j]	0.80[j]	1.10[j]	–	72.80[j]	–	–	–	–
Anthracite	5.00[j]	1.10[j]	2.20[j]	1.40[j]	1.80[j]	–	69.90[j]	0.02[k]	0.11[k]	–	–

Notes: – means no data, and the lowercase letters refer to the data source, as follows: a, Cao et al. (2008); b, Wang et al. (2009); c, Zhang et al. (2000); d, Zhang et al. (2008); e, Andreae and Merlet (2001); f, Tang et al. (2014); g, Tian et al. (2011); h, MEE (2014); i, Wei et al. (2008); j, Ministry of Environmental Protection (2016); and k, Tian et al. (2017)；When there are more than one data source, the average value from all data sources are used in this paper. PM, particulate matter; VOCs, volatile organic compounds; EC, elemental carbon; OC, organic carbon

region of Northeast China were invited to fill the questionnaire. However, during our survey process, we found that most student could not tell the amount of biomass of coal they burnt daily, or even annually, but they knew exactly the number acres of each fuel type they used. Therefore, in January 2018, we changed questionnaire to ask for the number acres of each biomass burnt at home every year, rather than the amount of biomass burnt. The survey method changed from paper questionnaire to online survey by Questionnaire Star (www.wjx.cn, an online questionnaire website) to involve more rural region people. Then farmers in the rural region of Northeastern China were invited to fill questionnaire online to ensure the accuracy of the investigation. Furthermore, more detail information about domestic biomass burning were collected, such as crop type, planting area of each crop, harvesting time, burning time, during time of burning, number acres of each biomass burnt at home, comprehensive utilization of crop residue, and even the location (town) of the farmers. In total, 432 effective questionnaires were collected from the different rural towns of Northeast China. With this information, we then calculated the yield of each biomass (corn, rice and soybean) per acre according to the statistical bureaus in Liaoning, Jilin, Heilongjiang provinces and four cities in the eastern Inner Mongolia, respectively. Finally, the amount of biomass burnt per household could be calculated by multiplying the number of acres of each biomass burnt by each household and the yield of each biomass type for its respective province.

The total amount of domestic burning in rural areas can be expressed as follows:

$$M = \sum H_i \times G_i \quad (2)$$

where M is the amount of biomass or coal annually burned across all rural areas of Northeast China (kg/yr), i sums across all provinces, H_i is the number of households in each province, and G_i is the average amount of biomass or coal burned in every household of a province (kg/yr). The number of households in each town is calculated according to the population of each town in rural region and the average person in each household. The population in each town published annually in the China Rural Statistical Yearbook (Department of Rural Social and Economic Investigation, National Bureau of Statistics, 2016). The values of G are collected by questionnaires, and presented in Table 2.

Table 2 The amount of biomass and coal domestic burned annually in each region of Northeast China (kg/(yr·household))

Province/Region	Biomass			Coal		
	Rice	Corn	Soybean	Bituminite	Briquette	Anthracite
Four cities in the eastern Inner Mongolia	104.47	3696.55	203.17	1522.50	480.75	496.75
Liaoning	800.74	2958.39	46.40	1344.88	424.66	438.80
Jilin	878.79	3667.01	54.69	874.65	276.18	285.37
Heilongjiang	1519.19	2343.47	341.00	1269.77	400.95	414.29

2.2.4 Spatiotemporal variations of pollutants from domestic burning

In order to get the high resolution spatial variations of domestic burning, and to provide grid based input data for air quality model simulation and forecasting, the domestic burning inventory is assigned into a 27 km × 27 km spatial grid using GIS software. The emission amount of a particular pollutant in a grid (emission intensity) also analyzed to obtained more detail spatial distribution information. For temporal distribution, an hourly distribution was defined based on our survey questionnaire. In this questionnaire we divided the year into the heating season (from October to April) and the non-heating season (from May to September), as cooking times and duration are different between the two seasons. Therefore, the survey questionnaire allowed for collection of data on start and end time of heating, start and end time of cooking for breakfast, lunch and dinner, duration of each heating and cooking in each province, split by these two seasons. Then the number of burning (both for heating and cooking) in each hour was counted based on survey questionnaire. Finally the hourly fraction was calculated by dividing the burning number of whole day by each hour.

3 Results

3.1 Domestic biomass burning emissions in rural region of Northeast China

The annual total domestic biomass burning emissions in 2016 of SO_2, NO_X, PM_{10}, $PM_{2.5}$, NH_3, CO, EC, OC, CO_2, CH_4 and VOCs (Volatile Organic Compounds) in rural Northeast China are 82.35 Gg, 135.35 Gg, 547.26 Gg, 508.83 Gg, 50.14 Gg, 6138.05 Gg, 61.94 Gg, 164.73 Gg, 104 395.30 Gg, 316.08 Gg and 574.79 Gg, respectively. Total emissions of particulate matter and gaseous pollutants for each city of Northeast China are presented in Table 3. These results indicate that Harbin, Suihua, Changchun, Qiqihar, Chifeng and Siping are the largest contributors; most of these cities are in Heilongjiang and Jilin provinces. In Harbin city, the annual emissions of SO_2, NO_X, PM_{10}, $PM_{2.5}$, NH_3, CO, EC, OC, CO_2, CH_4 and VOCs from domestic biomass burning are 5.74 Gg, 10.42 Gg, 43.52 Gg, 40.46 Gg, 4.05 Gg, 489.14 Gg, 4.50 Gg, 13.00 Gg, 7968.98 Gg, 26.50 Gg, and 47.21 Gg, respectively, with the total emissions contribution from all pollutants contributing to more than 7% of the total emissions from Northeast China. Each city, however, has a different emissions profile across the various pollutants. For SO_2 and EC, the major contributor is Changchun, followed by Harbin, Suihua, Chifeng and Qiqihar, while for other pollutants, the major contributor is Harbin, followed by Suihua, Changchun, Qiqihar and Chifeng. We posit that this may be due to using different major crop straws.

In addition to determining the total emissions from domestic biomass burning in rural areas, as discussed above, this study also investigated the emissions intensity of particulate matter and gaseous pollutants in these areas (Fig. 2). The results show high emission intensities for all pollutants originating along the Harbin-Changchun-Shenyang City Cluster. In Liaoning Province, the emissions intensity was not significantly different between cities, and was high for all cities in this province. In Jilin Province, the central region showed high emissions intensity (Yushu, Fuyu, Dehui, Changchun), whereas the western and eastern regions exhibited low emissions intensities. In Heilongjiang Province, high emissions intensities were concentrated around the southern region (Harbin, Daqing, Suihua) and dispersed throughout the northeastern region (the Sanjiang Plain) of the province. Conversely, nearly no emissions were seen in the northeastern Inner Mongolia or the northwestern Heilongjiang. These emissions intensities are consistent with the distribution of population and agriculture seen throughout these regions, which is not unexpected given that denser populations are correlated with higher levels of combustion of biomass, and therefore, resultant emissions. Indeed, high emissions were associated with greater agricultural and rural activities (Zhou et al., 2017).

3.2 Domestic coal burning emission in rural region of Northeast China

The total annual domestic coal burning emissions of SO_2, NO_X, CO, VOCs, PM_{10} and $PM_{2.5}$ in rural Northeast China in 2016 are 255.34 Gg, 50.51 Gg, 4246.05 Gg, 112.71 Gg, 332.68 Gg and 262.88 Gg, respectively. Heilongjiang and Liaoning provinces are the major contributors to all pollutants, following by Jilin Province and four cities in the eastern Inner Mongolia Autonomous region. This is related to heating duration and population in each province. In Harbin, the provincial capital of Heilongjiang Province, the heating season

Table 3 Total emissions of particulate matter and gaseous pollutants from domestic biomass burning in each city of Northeast China (Gg)

Region	SO_2	NO_X	PM_{10}	$PM_{2.5}$	NH_3	CO	EC	OC	CO_2	CH_4	VOCs
Chifeng	4.62	6.45	26.95	25.06	2.58	299.78	3.34	8.17	5306.27	14.74	27.14
Tongliao	3.27	4.56	19.06	17.72	1.82	211.97	2.36	5.78	3751.93	10.42	19.19
Hulun Beier	1.32	1.84	7.69	7.15	0.74	85.52	0.95	2.33	1513.74	4.21	7.74
Xinganmeng	1.79	2.51	10.47	9.73	1.00	116.47	1.30	3.18	2061.54	5.73	10.54
Shenyang	2.02	3.28	12.92	12.01	1.16	145.03	1.50	3.90	2504.28	7.31	13.44
Dalian	1.94	3.15	12.40	11.53	1.11	139.18	1.44	3.74	2403.30	7.02	12.90
Anshan	1.66	2.70	10.62	9.88	0.95	119.24	1.24	3.21	2058.91	6.01	11.05
Fushun	1.07	1.73	6.82	6.34	0.61	76.55	0.79	2.06	1321.89	3.86	7.10
Benxi	0.52	0.84	3.30	3.07	0.30	37.06	0.38	1.00	640.01	1.87	3.44
Dandong	1.78	2.89	11.38	10.58	1.02	127.77	1.32	3.44	2206.29	6.44	11.84
Jinzhou	2.17	3.53	13.88	12.91	1.25	155.84	1.62	4.19	2690.96	7.86	14.45
Yingkou	1.34	2.18	8.59	7.99	0.77	96.42	1.00	2.59	1664.96	4.86	8.94
Fuxin	1.50	2.44	9.60	8.93	0.86	107.75	1.12	2.90	1860.65	5.43	9.99
Liaoyang	1.37	2.22	8.74	8.13	0.78	98.16	1.02	2.64	1694.99	4.95	9.10
Panjin	0.70	1.13	4.47	4.15	0.40	50.14	0.52	1.35	865.87	2.53	4.65
Tieling	2.47	4.01	15.80	14.69	1.42	177.32	1.84	4.77	3061.94	8.94	16.44
Chaoyang	3.08	5.00	19.69	18.30	1.77	220.98	2.29	5.94	3815.74	11.14	20.48
Huludao	2.59	4.21	16.57	15.41	1.49	186.00	1.93	5.00	3211.80	9.38	17.24
Changchun	6.30	10.09	39.76	36.97	3.58	446.01	4.67	12.01	7733.67	22.38	41.20
Jilin	3.10	4.96	19.55	18.17	1.76	219.25	2.30	5.90	3801.70	11.00	20.25
Siping	3.44	5.50	21.68	20.15	1.95	243.15	2.55	6.55	4216.25	12.20	22.46
Liaoyuan	1.15	1.84	7.26	6.75	0.65	81.39	0.85	2.19	1411.32	4.08	7.52
Tonghua	1.93	3.09	12.20	11.34	1.10	136.80	1.43	3.68	2372.16	6.86	12.64
Baishan	0.71	1.14	4.51	4.19	0.41	50.54	0.53	1.36	876.30	2.54	4.67
Songyuan	3.38	5.41	21.34	19.84	1.92	239.35	2.51	6.44	4150.28	12.01	22.11
Baicheng	1.86	2.98	11.77	10.94	1.06	132.00	1.38	3.55	2288.86	6.62	12.19
Yanbianzhou	0.97	1.55	6.12	5.69	0.55	68.62	0.72	1.85	1189.82	3.44	6.34
Harbin	5.74	10.42	43.52	40.46	4.05	489.14	4.50	13.00	7968.98	26.53	47.21
Qiqihar	4.38	7.96	33.22	30.89	3.09	373.41	3.44	9.92	6083.48	20.25	36.04
Jixi	0.91	1.65	6.88	6.40	0.64	77.39	0.71	2.06	1260.82	4.20	7.47
Hegang	0.24	0.43	1.80	1.67	0.17	20.23	0.19	0.54	329.65	1.10	1.95
Shuangyashan	0.58	1.06	4.42	4.11	0.41	49.71	0.46	1.32	809.80	2.70	4.80
Daqing	1.80	3.27	13.64	12.69	1.27	153.36	1.41	4.08	2498.46	8.32	14.80
Yichun	0.29	0.52	2.16	2.01	0.20	24.29	0.22	0.65	395.80	1.32	2.34
Jiamusi	1.71	3.10	12.94	12.04	1.20	145.50	1.34	3.87	2370.43	7.89	14.04
Qitaihe	0.38	0.68	2.84	2.64	0.26	31.97	0.29	0.85	520.91	1.73	3.09
Mudanjiang	1.47	2.66	11.12	10.34	1.03	124.99	1.15	3.32	2036.35	6.78	12.06
Heihe	0.96	1.75	7.29	6.78	0.68	81.93	0.75	2.18	1334.73	4.44	7.91
Suihua	5.74	10.41	43.47	40.42	4.04	488.60	4.50	12.99	7960.24	26.50	47.15
Da Hinggan Ling	0.11	0.20	0.82	0.76	0.08	9.22	0.08	0.25	150.23	0.50	0.89
Total	82.35	135.35	547.26	508.83	50.14	6138.05	61.94	164.73	104395.30	316.08	574.79

Notes: PM, particulate matter; VOCs, volatile organic compounds; EC, elemental carbon; OC, organic carbon

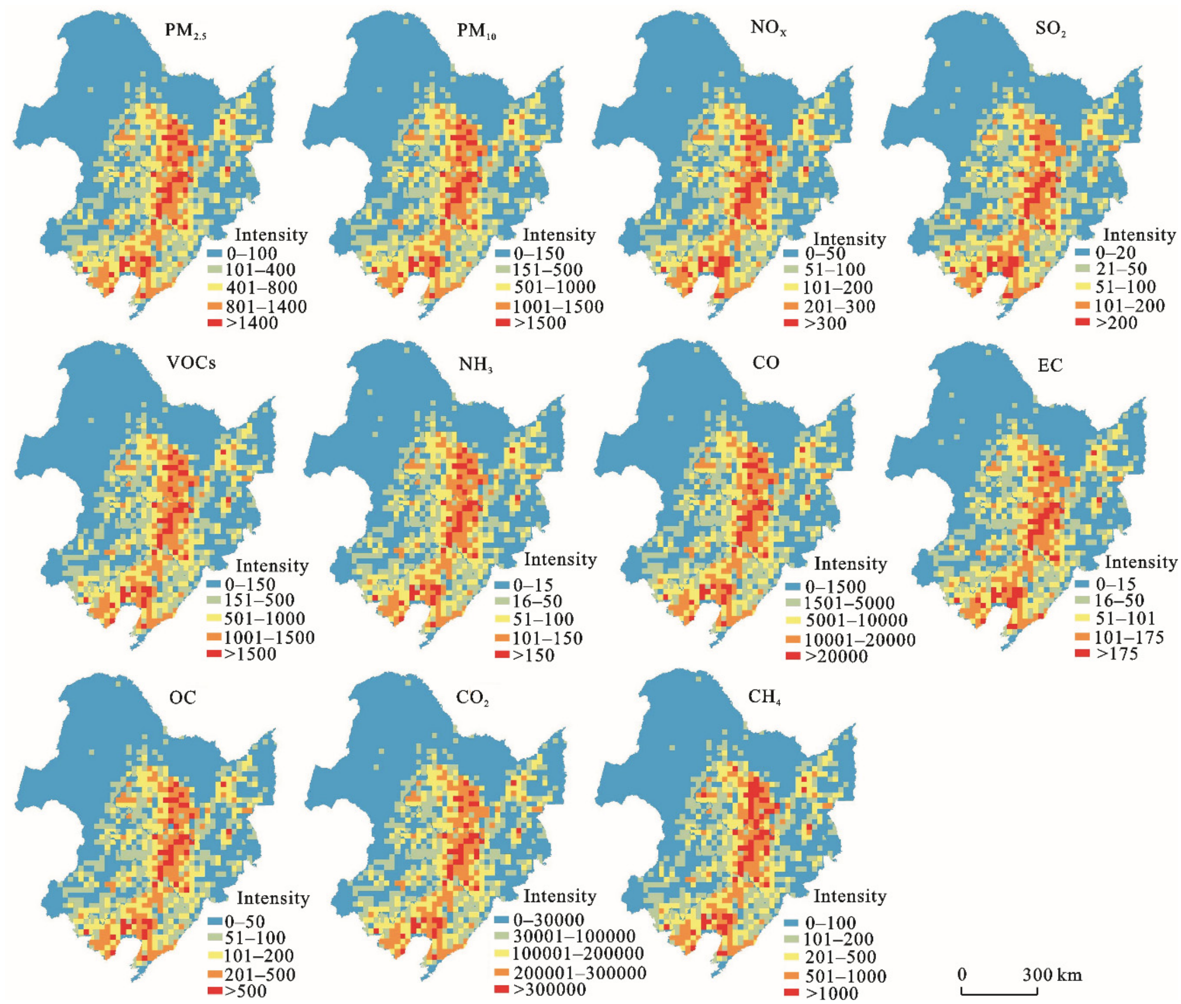

Fig. 2 Emission intensity (Mg/grid) of particulate matter and gaseous pollutants from domestic biomass burning in rural region of Northeast China. PM, particulate matter; VOCs, volatile organic compounds; EC, elemental carbon; OC, organic carbon

usually lasts from 15 October to 15 April, which is started earlier and ended later about 20 d than other cities in Jilin and Liaoning provinces. While the population in Liaoning Province is greater than other provinces in Northeast China. The prolonged heating season and greater population caused more coal consumption for heating in Heilongjiang and Liaoning provinces. The contributions of domestic coal burning to $PM_{2.5}$ from Heilongjiang, Liaoning, Jilin provinces and the four cities in the eastern Inner Mongolia autonomous region to the region were 34.6%, 33.7%, 16.9% and 14.8%, respectively. Total domestic coal burning emissions of particulate matter and gaseous pollutants for each city of Northeast China are given in Fig. 3. These results suggest that Harbin, Suihua, Qiqihar and Chifeng are the largest city contributors, collectively contributing more than 28% of the region's total emissions. Conversely, Da Hinggan Ling Prefecture, Hegang and Yichun contributed the least, with their total contribution to the region being less than 1 %. These results were also reflected in those for domestic biomass burning, with the Harbin-Changchun-Shenyang City Cluster exhibiting higher emissions intensities for all pollutants. This is, again, due to the fact that population density is the predominant factor determining domestic burning emissions.

Contributions from different coal types burned throughout the rural areas of Northeast China were also examined (Fig. 4). The results revealed that bituminite is the major contributor, accounting for, on average, about 80% of the total emissions for all pollutants. This is consistent with patterns of coal consumption; in Northeast China, the annual consumption of domestic coal burning for bituminite, briquette and anthracite are 22.84 Gg, 7.21 Gg and 7.45 Gg, respectively. Moreover,

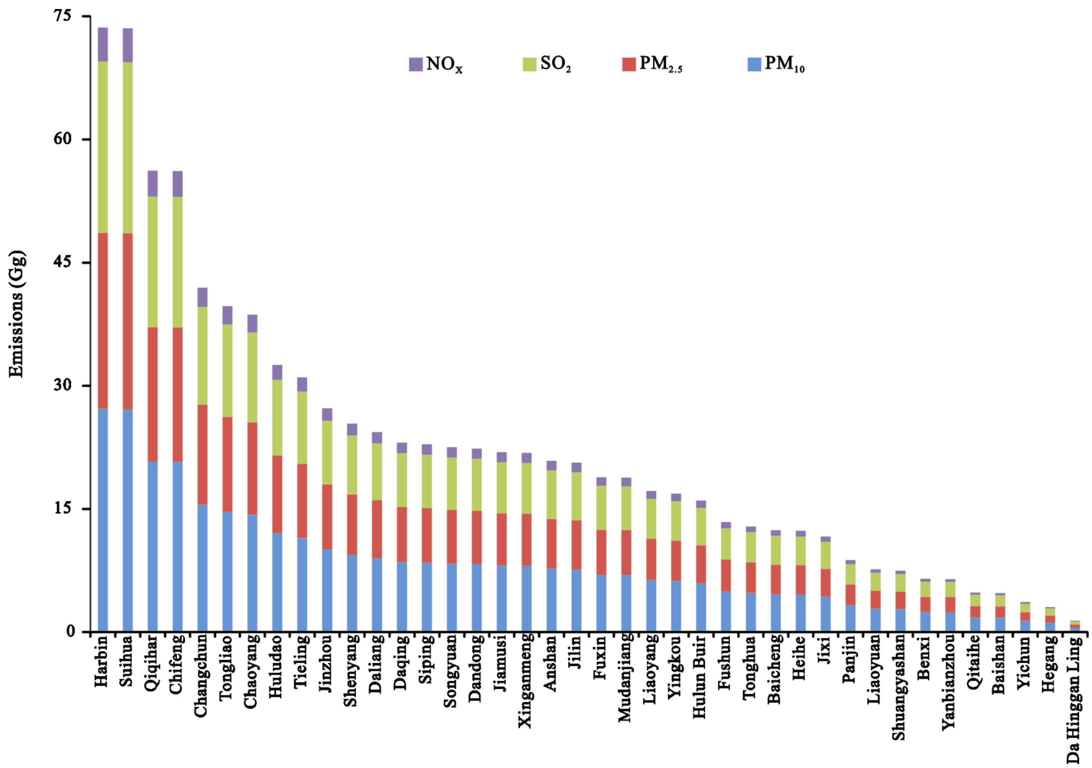

Fig. 3 Emissions of SO_2, NO_X, $PM_{2.5}$ and PM_{10} from domestic coal burning in Northeast China. PM, particulate matter

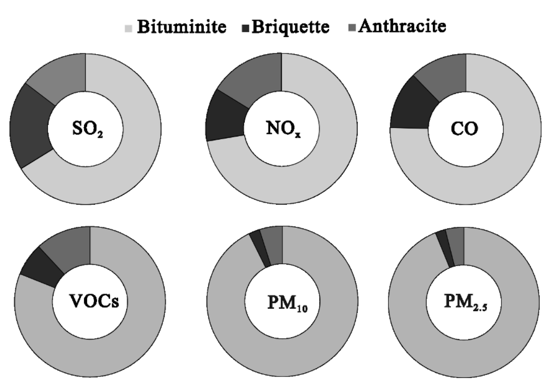

Fig. 4 Contribution of different coal to particulate matter and gaseous pollutants in rural region of Northeast China. PM, particulate matter; VOCs, volatile organic compounds

contributions to various pollutants from each coal type are analyzed. For all the six pollutants (SO_2, NO_X, CO, VOCs, PM_{10} and $PM_{2.5}$) the major contributor is bituminite, especially for PM, with a contribution of more than 90%. While briquette and anthracite shown similar contribution to all pollutants. This is associated with burnt amounts and emission factors. The annual consumption of bituminite is about three times greater than briquette and anthracite, during to the lower price than other coal types. Furthermore, the emission factors for all pollutants from bituminite are also significant greater than other two coal types.

3.3 Total domestic burning emission in rural Northeast China

The total domestic burning emissions of SO_2, NO_X, CO, VOCs, PM_{10} and $PM_{2.5}$ in rural Northeast China in 2016 are 337.69 Gg, 185.87 Gg, 13 384.10 Gg, 682.49 Gg, 879.94 Gg and 771.71 Gg, respectively. Contributions to various pollutants from coal and biomass burning are different (Fig. 5). For example, biomass burning was the major source of the majority of pollutants, e.g., PM, VOCs, NO_X and CO. For VOCs emissions, biomass and coal contributed 83.3% and 16.7% to total burning, respectively. For PM, NO_X and CO emissions, biomass burning accounted for 59.1% to 72.6% of total burning emissions. In contrast, however, coal burning was the major source of SO_2 emissions, with a contribution of greater than 75% (meaning that biomass burning accounted for less than 25%). These results suggest that to control PM and VOCs emissions from domestic burning, the burning of biomass should be taken into account, whereas to reduce SO_2 emissions, domestic coal burning should be considered.

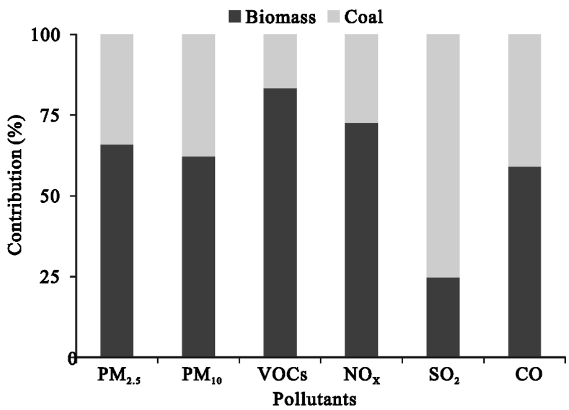

Fig. 5 The contribution of domestic biomass and coal burning to pollutant emissions in rural region of Northeast China. PM, particulate matter; VOCs, volatile organic compounds

The spatial distribution and emissions intensity of domestic biomass and coal burning were also investigated in this study. All gaseous pollutants and particulate matters presented similar emissions intensity distributions. In Fig. 6, we have taken $PM_{2.5}$ and VOCs as examples to analyze the spatial missions distribution. The 27 km × 27 km grid emissions distribution reveals that the Harbin-Changchun-Shenyang City Cluster is responsible for the highest emissions intensities in the region. These high emissions intensities are concentrated on the cities of Harbin, Suihua, Changchun, Qiqihar, and Chifeng, with emissions intensities for $PM_{2.5}$ and VOCs of more than 2000 Mg per grid cell. Especially during the Spring Festival, due to the population of rural counties increased, domestic burning and pollutants emissions increased, and adverse weather conditions (especially radiation inversion and urban-rural heat island effects), both urban and rural areas were suffered from regional haze. Low emission intensities are mainly distributed in the north of Heilongjiang Province, and the northeast of the eastern Inner Mongolia and the eastren part of of Jilin. These regions are covered by forests, mountains and rivers, and with low population densities, result in lower emissions.

3.4 Temporal variation of domestic burning in rural Northeast China

Previous studies have demonstrated that domestic fuel consumption is strongly dependent on season, and that the daily temporal variation of domestic burning varies throughout the year (Li et al., 2017), a conclusion that is corroborated by this study. Both during the heating and non-heating seasons, there are three burning peaks throughout the day (6−7 am, 12 pm and 4−7 pm) (Fig. 7); these burning peaks coincide with breakfast, lunch and dinner times. The burning peaks, however, are more notable throughout the non-heating season than the heating season, reflecting living habits in Northeast China. During the heating season, people need to burn fuel for heating throughout the day, particularly in the early morning and late nights (and less so during cooking times). There are, therefore, consistent emissions throughout the day, with only small peaks over cooking times. The contribution of cooking at 7 am, 1 pm and 5 pm to total daily burning is less than 30%. This suggests that heating is the major cause of burning during the

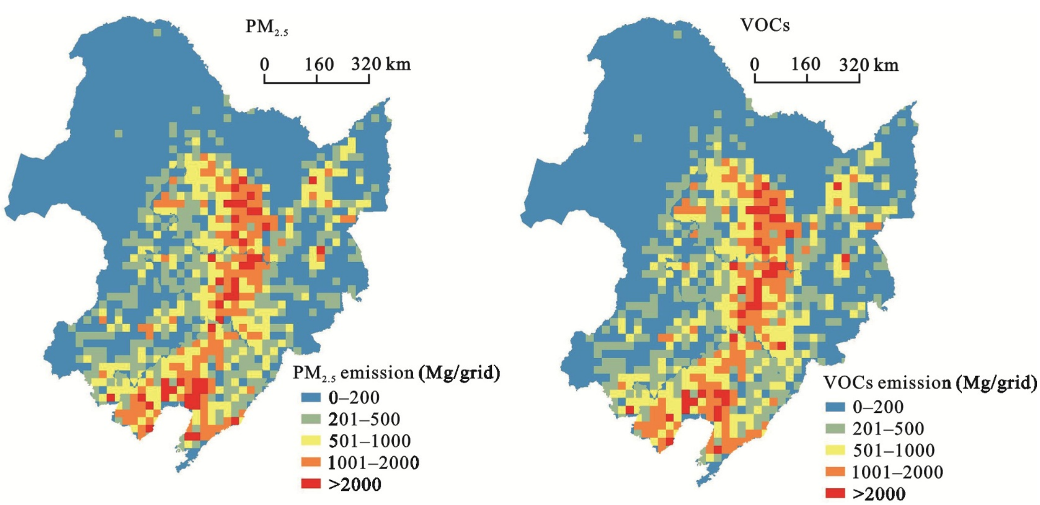

Fig. 6 Emissions intensity of $PM_{2.5}$ and VOCs from domestic biomass and coal burning in rural Northeast China (Mg/grid). PM, particulate matter; VOCs, volatile organic compounds

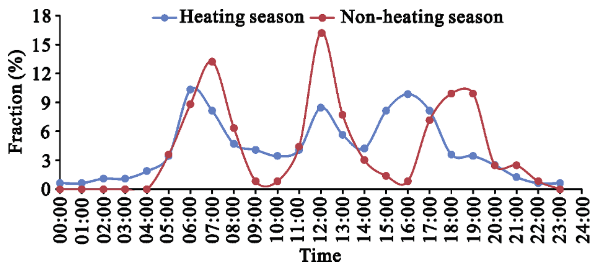

Fig. 7 The mean diurnal variation of domestic burning during heating and non-heating season in rural of Northeast China

heating season. During the non-heating season, however, the only burning that occurs is due to cooking: at 7 am, people prepare breakfast, and there is a corresponding burning peak. Likewise, there are burning peaks for lunch and dinner cooking at 12 pm and 6−7 pm. The contribution of burning at 7 am, 12 pm and 6−7 pm is approximately 50% of the daily total.

4 Discussion

Exposure to household air pollution from heating and cooking with solid fuels is a major health risk. Emission inventory of domestic burning of solid fuels is needed for risk assessment and policy implication. Globally, the proportion of households relying on solid fuels for cooking has decreased from 62% in 1980 to 41% in 2010 (Bonjour et al., 2013). However, the number of people exposed to household air pollution has remained stable with the growth of population. Particular in Africa and Southeast Asia, where more than 60% of households using solid fuels, while in Americas and Europe, the proportion is less than 20% (Bonjour et al., 2013). On the other hand, the research about solid fuels mainly focus on developed county, with limited studied conducted in developing county. According to the first Chinese Environmental Exposure-Related Human Activity Patterns Survey, solid fuels were widely used for cooking and heating in China. Especially in rural region, solid fuels such as biomass and coal were the predominant fuel used for cooking (Duan et al., 2014). However the emission from biomass and coal burning indoor with high spatiotemporal resolution is limited, which is unfavorable for risk assessment of exposure to household air pollution.

In this study, we developed a town-level emission inventory of domestic biomass and coal burning in rural region based on household consumption. The results revealed that biomass burning was the major domestic burning source in rural region of Northeast China, especially in Heilongjiang Province. This result is consistent with previous studies (Li et al., 2017; Tian et al., 2017), as crop residue is more easily available and cheaper than coal fuel in the main grain production areas. A previous study has also illustrated that Heilongjiang Province contributed the most agricultural biomass burning emission in China (Tian et al., 2017). Heilongjiang and Jilin provinces are China's major grain producing regions. Being free and available, crop straw has become the main household fuel in these regions, which need large amounts of fuel for continual heating during the winter months (Xing et al., 2018). Furthermore, the Harbin-Changchun-Shenyang city cluster is responsible for the highest emissions intensities in the region. These emissions intensities are consistent with the distribution of population and agriculture seen throughout these regions, which is not unexpected given that denser populations are correlated with higher levels of combustion of biomass, and therefore, resultant emissions. Indeed, high emissions were associated with greater agricultural and rural activities (Zhou et al., 2017). While low emission intensities are mainly distributed in the north of Heilongjiang Province, and the northeast of the eastern Inner Mongolia and east of Jilin. These regions are covered by forests, mountains and rivers, and with low population densities, result in lower emissions. High concentrations of air pollutants in southern and central regions while lower values in surrounding regions of Northeast China also was documented by Li et al. (2019b). Finally, high emission of PM and VOCs were found in high population density region. A previous study conducted in Xi'an in Northwest China also revealed that PM pollution in rural areas was caused by intense indoor coal combustion for heating (Dai et al., 2019). Another study about the impacts of heating emissions on the environment and human health in North China also documented that the environment in Heilongjiang province was severely affected by heating, due to cold weather and large heating consumption (Li et al., 2019a). Furthermore, the city of Qiqihar was the most severely impacted city caused by heating emissions (Li et al., 2019a). These high emissions areas are the major agricultural regions with abundant farms, and additionally, high population densities.

A comprehensive assessment of solid fuels used in household would help provide a better understanding of the impact of indoor air pollution on human health and atmospheric environment. Exposure to household air pollution is one of the most important causes of illness for women and child in developing county. Despite the emission inventory of all kinds of gaseous pollutants and particular matter, the number of persons exposed to indoor air pollution should be estimated. Particularly in the rural of Northeast China, the severely cold climate made people spend most of time stay at home during the heating season. The daily temporal variation of domestic burning in the heating season suggested that solid fuels burning occurred each hour. It is urgent to change the solid fuels to cleaner fuels to improve indoor air quality and reduce the impact on human health.

5 Conclusions

Domestic burning, a major anthropogenic source of ambient air pollution, has been relatively under acknowledged in air pollution control strategies. High-resolution emissions inventories of domestic burning are scarce, even in areas where these emissions are high, such as Northeast China. In this study, we used household consumption data to develop a town-level domestic biomass and coal burning emissions inventory for rural Northeast China, with hourly temporal resolution. Total domestic burning emissions of SO_2, NO_X, CO, VOCs, PM_{10} and $PM_{2.5}$ in rural Northeast China are 337.69 Gg, 185.87 Gg, 13 384.10 Gg, 682.49 Gg, 879.94 Gg and 771.71 Gg, respectively. High emissions intensities were concentrated around Harbin, Suihua, Changchun, Qiqihar, and Chifeng, each with an emission intensity for $PM_{2.5}$ and VOCs of more than 2000 Mg per grid cell. Additionally, there are three burning peaks that occur both in the heating and non-heating seasons; however, the burning peaks during the non-heating season are more notable than those in the heating season. This high-resolution emissions inventory provides detailed information that could be utilized in future quantitative studies using air quality models, such as estimating the impact of domestic burning on indoor air quality and residents' health in rural regions. The high temporal resolution of the data will assist in understanding the temporal impact of the residential solid fuel combustion on local ambient air.

The study provide valuable data about domestic biomass and coal burning for cooking and heating in rural region of Northeast China. To some extent, it validated the estimation of solid fuels at the regional even town level. One limitation of this study is that most household used solid fuels for cooking and heating not only in rural region but also in urban region. More detailed domestic burning information in urban region should be considered in future to obtain the comprehensive emission. The results of this study have revealed that biomass is the major residential fuel in rural Northeast China, and as such, domestic biomass burning emissions deserve more attention when considering controls for indoor air pollution and regional haze. In addition, comprehensive utilization of crop residues should be considered to reduce domestic burning emissions. Furthermore, cleaner fuels such as smokeless coal should be considered in rural region to improve indoor air quality and reduce the impact on human health.

References

Andreae M O, Merlet P, 2001. Emission of trace gases and aerosols from biomass burning. *Global Biogeochemical Cycles*, 15(4): 955–966. doi: 10.1029/2000GB001382

Badamassi A, Xu D Y, Leyla B H, 2017. The impact of residential combustion emissions on health expenditures: empirical evidence from Sub-Saharan Africa. *Atmosphere*, 8(9): 157. doi: 10.3390/atmos8090157

Bonjour B, Adair-Rohani H, Wolf J et al., 2013. Solid fuel use for household cooking: country and regional estimates for 1980–2010. *Environmental Health Perspectives*, 121(7): 784–790. doi: 10.1289/ehp.1205987

Cao G L, Zhang X Y, Gong S L et al., 2008. Investigation on emission factors of particulate matter and gaseous pollutants from crop residue burning. *Journal of Environmental Sciences*, 20(1): 50–55. doi: 10.1016/S1001-0742(08)60007-8

Chen J M, Li C L, Ristovski Z et al., 2017. A review of biomass burning: emissions and impacts on air quality, health and climate in China. *Science of the Total Environment*, 579: 1000–1034. doi: 10.1016/j.scitotenv.2016.11.025

Chen Weiwei, Zhang Shichun, Tong Quansong et al., 2018a. Regional characteristics and causes of haze events in northeast China. *Chinese Geographical Science*, 28(5): 836–850. doi: 10.1007/s11769-018-0965-3

Chen Y L, Shen H Z, Smith K R et al., 2018b. Estimating household air pollution exposures and health impacts from space heating in rural China. *Environment International*, 119: 117–124. doi: 10.1016/j.envint.2018.04.054

Cheng M M, Zhi G R, Tang W et al., 2017. Air pollutant emission from the underestimated households' coal consumption source in China. *Science of the Total Environment*, 580: 641–650. doi: 10.1016/j.scitotenv.2016.12.143

Dai Q L, Bi X H, Song W B et al., 2019. Residential coal combustion as a source of primary sulfate in Xi'an, China. *Atmospheric Environment*, 196: 66–76. doi: 10.1016/j.atmosenv.2018.10.00.

Department of Rural Social and Economic Investigation, National Bureau of Statistics, 2016. *China Rural Statistical Yearbook 2016*. Beijing: China Statistics Press. (in Chinese)

Desai M A, Mehta S, Smith K, 2004. *Indoor Smoke from Solid Fuels: Assessing the Environmental Burden of Disease at National and Local Levels*. Geneva: World Health Organization

Du W, Li X Y, Chen Y C et al., 2018. Household air pollution and personal exposure to air pollutants in rural China – A review. *Environmental Pollution*, 237: 625–638. doi: 10.1016/j.envpol.2018.02.054

Duan X L, Jiang Y, Wang B B et al., 2014. Household fuel use for cooking and heating in China: results from the first Chinese Environmental Exposure-Related Human Activity Patterns Survey (CEERHAPS). *Applied Energy*, 136: 692–703. doi: 10.1016/j.apenergy.2014.09.066

Fleming L T, Lin P, Laskin A et al., 2018. Molecular composition of particulate matter emissions from dung and brushwood burning household cookstoves in Haryana, India. *Atmospheric Chemistry and Physics*, 18(4): 2461–2480. doi: 10.5194/acp-18-2461-2018

Huang W, Baumgartner J, Zhang Y X et al., 2015. Source apportionment of air pollution exposures of rural Chinese women cooking with biomass fuels. *Atmospheric Environment*, 104: 79–87. doi: 10.1016/j.atmosenv.2014.12.066

Jiang R T, Bell M L, 2008. A comparison of particulate matter from biomass-burning rural and non-biomass-burning urban households in Northeastern China. *Environmental Health Perspectives*, 116(7): 898–906. doi: 10.1289/ehp.10622

Li H, You S J, Zhang H et al., 2019a. Analysis of the impacts of heating emissions on the environment and human health in North China. *Journal of Cleaner Production*, 207: 728–742. doi: 10.1016/j.jclepro.2018.10.013

Li Q, Jiang J K, Cai S Y et al., 2016. Gaseous ammonia emissions from coal and biomass combustion in household stoves with different combustion efficiencies. *Environmental Science & Technology Letters*, 3(3): 98–103. doi: 10.1021/acs.estlett.6b00013

Li Q, Jiang J K, Wang S X et al., 2017. Impacts of household coal and biomass combustion on indoor and ambient air quality in China: current status and implication. *Science of the Total Environment*, 576: 347–361. doi: 10.1016/j.scitotenv.2016.10.080

Li Xiaolan, Hu Xiaoming, Shi Shuaiyi et al., 2019b. Spatiotemporal variations and regional transport of air pollutants in two urban agglomerations in Northeast China plain. *Chinese Geographical Science*, 29(6): 917–933. doi: 10.1007/s11769-019-1081-8

Liu J, Mauzerall D L, Chen Q et al., 2016. Air pollutant emissions from Chinese households: a major and underappreciated ambient pollution source. *Proceedings of the National Academy of Sciences of the United States of America*, 113(28): 7756–7761. doi: 10.1073/pnas.1604537113

MEE, 2014. Guide for compiling atmospheric pollutant emission inventory for biomass burning, Ministry of Ecology and Environment of the People's Republic of China. Available at http://www.mee.gov.cn/gkml/hbb/bgg/201501/t20150107_293955.htm. 2019-08-20. (in Chinese)

Mestl H ES, Aunan K, Seip H M et al., 2007. Urban and rural exposure to indoor air pollution from domestic biomass and coal burning across China. *Science of the Total Environment*, 377(1): 12–26. doi: 10.1016/j.scitotenv.2007.01.087

Ministry of Environmental Protection, 2016. Guide for compiling atmospheric pollutant emission inventory for residential coal burning, Ministry of Ecology and Environment of the People's Republic of China. Available at http://www.mee.gov.cn/gkml/hbb/bgg/201610/t20161031_366528.htm. 2019-08-20. (in Chinese)

Shen G F, Wei S Y, Wei W et al., 2012. Emission factors, size distributions, and emission inventories of carbonaceous particulate matter from residential wood combustion in rural China. *Environmental Science & Technology*, 46(7): 4207–4214. doi: 10.1021/es203957u

Shen G F, Chen Y C, Xue C Y et al., 2015. Pollutant emissions from improved coal- and wood-fuelled cookstoves in rural households. *Environmental Science & Technology*, 49(11): 6590–6598. doi: 10.1021/es506343z

Tang Xibin, Huang Cheng, Lou Shengrong et al., 2014. Emission factors and pm chemical composition study of biomass burning in the Yangtze river delta region. *Environmental Science*, 35(5): 1623–1632. (in Chinese)

Tian Hezhong, Zhao Dan, Wang Yan, 2011. Emission inventories of atmospheric pollutants discharged from biomass burning in China. *Acta Scientiae Circumstantiae*, 31(2): 349–357. (in Chinese)

Tian J, Ni H Y, Cao J J et al., 2017. Characteristics of carbonaceous particles from residential coal combustion and agricultural biomass burning in China. *Atmospheric Pollution Research*, 8(3): 521–527. doi: 10.1016/j.apr.2016.12.006

Wang R, Tao S, Wang W T et al., 2012. Black carbon emissions in China from 1949 to 2050. *Environmental Science & Technology*, 46(14): 7595−7603. doi: 10.1021/es3003684

Wang S X, Wei W, Du L et al., 2009. Characteristics of gaseous pollutants from biofuel-stoves in rural China. *Atmospheric Environment*, 43(27): 4148–4154, doi: 10.1016/j.atmosenv.2009.05.040

Wei W, Wang S X, Chatani S et al., 2008. Emission and speciation of non-methane volatile organic compounds from anthropogenic sources in China. *Atmospheric Environment*, 42(20): 4976–4988. doi: 10.1016/j.atmosenv.2008.02.044

Wen Xin, Zhang Pingyu, Liu Daqian, 2018. Spatiotemporal variations and influencing factors analysis of $PM_{2.5}$ concentrations in Jilin Province, Northeast China. *Chinese Geographical Science*, 28(5): 810–822. doi: 10.1007/s11769-018-0992-0

WHO, 2018. Household air pollution and health. Available at https://www.who.int/news-room/fact-sheets/detail/household-air-pollution-and-health/. Cited 5 July 2020

Wilnhammer M, Wittkopf S, Richter K et al., 2017. The impact of a new emission control act on particulate matter emissions from residential wood energy use in Bavaria, Germany. *Journal of Cleaner Production*, 145: 134–141. doi: 10.1016/j.jclepro.2017.01.039

Xing X F, Zhou Y, Lang J L et al., 2018. Spatiotemporal variation of domestic biomass burning emissions in rural China based on a new estimation of fuel consumption. *Science of the Total Environment*, 626: 274–286. doi: 10.1016/j.scitotenv.2018.01.048

Yadav I C, Devi N L, Li J et al., 2017. Biomass burning in Indo-China peninsula and its impacts on regional air quality and global climate change—a review. *Environmental Pollution*, 227: 414–427. doi: 10.1016/j.envpol.2017.04.085

Yang G Y, Zhao H M, Tong D Q et al., 2020. Impacts of post-harvest open biomass burning and burning ban policy on severe haze in the Northeastern China. *Science of the Total Environment*, 716: 136517. doi: 10.1016/j.scitotenv.2020.136517

Zhang H F, Ye X N, Cheng T T et al., 2008. A laboratory study of agricultural crop residue combustion in China: emission factors and emission inventory. *Atmospheric Environment*, 42(36): 8432–8441, doi: 10.1016/j.atmosenv.2008.08.015

Zhang J, Smith K R, Ma Y et al., 2000. Greenhouse gases and other airborne pollutants from household stoves in China: a database for emission factors. *Atmospheric Environment*, 34(26): 4537–4549, doi: 10.1016/s1352-2310(99)00450-1

Zhang J F, Smith K R, 2007. Household air pollution from coal and biomass fuels in China: measurements, health impacts, and interventions. *Environmental Health Perspectives*, 115(6): 848–855. doi: 10.1289/ehp.9479

Zhou Y, Xing X F, Lang J L et al., 2017. A comprehensive biomass burning emission inventory with high spatial and temporal resolution in China. *Atmospheric Chemistry and Physics*, 17(4): 2839–2864, doi: 10.5194/acp-17-2839-2017